Determination of Marine Aerosol Properties Using a Bistatic Nephelometer

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LONG-TERM GOALS

The long term goal is to characterize aerosols remotely in the marine atmospheric boundary layer (MABL) based on instruments and analytic methods that use angle-dependent polarization information from light scattered by aerosols. This information will enable accurate prediction of optical properties and light propagation in the MABL.

OBJECTIVES

The objective is to develop a new instrument and method to characterize atmospheric aerosols by remotely sensing scattered light. A bi-static nephelometer (an instrument with separately pointed light source and detector that probes at a distance) appears to be an effective tool to supply data regarding aerosol content and light propagation in the MABL. It provides a means to probe the area about its mounting platform whether it is mounted on-shore or shipboard. Light scattering techniques have the advantage of providing a direct, rapid *in situ* measurement of the optical properties of aerosols. A bi-static nephelometer using polarization modulation can probe at a distance to measure the polarization of light scattered from aerosols in the sensed region. A significant advantage inherent to polarization modulation is that it allows the use of an intensity normalization procedure that permits determination of aerosol size distributions and optical properties using polarization information independent of scattered light intensity. By using various scanning strategies, the aerosols in the MABL about the platform may be characterized for visibility and aerosol content in spatial detail (mapped) with time.

To use the bistatic nephelometer measurements effectively an analytical modeling effort that predicts the polarization properties of light scattering from typical atmospheric aerosols is necessary. Therefore scattering models based on Mie calculations must be supplemented with more general calculational approaches that provide for scattering from non-spherical particles. Therefore another objective is to develop viable models to calculate scattering from a range of regular and irregular non-spherical particles.

APPROACH

To characterize the intensity and polarization of light scattered from marine aerosols we modified our bi-static nephelometer [designed for use on sea ice] and are testing its operation under controlled conditions in preparation for field measurements. The bi-static nephelometer measures light scattering with an independently aimed light source and detector separated by a distance. The laser light source and detector can be directed to any region of space or scanned about the instrument platform (*eg*. ship bow and stern or two locations on shore). The technique provides direct, rapid *in situ* measurement of

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aerosol properties in the lower MABL and requires no collection thus eliminating potential sample perturbation. A method of intensity normalization using the time varying signals arising from polarization modulation provides data that enable determination of size distributions and optical properties independent of aerosol density. By using various strategies to scan the light source and detector independenly, the spatial and temporal character (4D) of aerosols in the MABL about the platform may be mapped. This information can be used to determine the most effective polarimetric strategy to improve visibility and target discrimination.

As polarization-dependent data become available for the MABL, particularly in the coastal zone, the analysis methods used to retrieve size distributions and optical properties of the scatterers from the measurements must be extended. The presence of non-spherical particles makes use of Mie calculations of scattering from spherical particles inappropriate. To predict scattering from non-spherical particles a computer code was developed using a coupled-dipole approximation (CDA). The approach allows the calculation of scattering from regular and irregular non-spherical shapes. However, realistic particle simulation requires building particles from a multiplicity of dipoles and then calculating all their interactions. This tends to be computationally intensive. To reduce the computational time, a parallel version of the CDA code was implemented on the LBNL 640-processor Cray T3E-900 (theoretical peak speed of 575 Gflops). The parallel codes permit simultaneous calculation of scattering from particles having different sizes, orientations, and shapes.

WORK COMPLETED

The bistatic nephelometer, shown schematically in Fig. 1 (a) and in cross section in Fig. 1 (b), uses polarization modulation to measure the elements of the Mueller matrix for the atmospheric aerosols. The beam is deflected by two back-to-back right angle prisms in such a way that the light re-emerges from this biprism "mirror" in the plane defined by the initial beam direction. The dotted line in Fig. 1 (a) indicates the scattering plane. A programmed stepper motor attached to the biprism mirror varies the angle of the deflected beam that is controlled by a lap-top computer. The beam then exits the instrument and enters the atmosphere. The scattered light is collected by a cylindrical plano-concave lens and passes through a second biprism mirror, also actuated by a stepper motor. The scattered light traverses the polarizing and laser band-pass filters before being detected by a photomultiplier tube. The detector optics have a ~ 2.5° half-width field of view in the scattering plane and a choice of larger apertures.

The ac components of the signal proportional to the matrix elements are detected synchronously with a lock-in amplifier. By choosing the proper combination of (1) initial polarization state and PEM position; (2) final polarizing filters; and (3) lock-in amplifier frequency or dc detection, measurements can be made of all 16 components of the Mueller matrix. The lap-top computer controls the operation of entire instrument and records the output of the lock-in amplifier and signal normalization information. The PMT can be operated at either a constant dc voltage or constant dc current mode. For most measurements the constant dc current mode is used. This mode provides automatic normalization of the ac signals from the lock-in amplifier as the laser and detector angles are scanned that are proportional to the chosen Mueller matrix element.

Nephelometer Operation

The bistatic nephelometer was modified for measurement of atmospheric aerosols. This involved the system, eliminating the index-matching ice lenses, remounting, and extensive modifications to the software controlling the stepper motors that move the scanning mirrors. The operation of the nephelometer was verified by laboratory measurements of well-characterized scattering systems. The measurements were performed in a water tank containing an aqueous suspension of latex spheres of known diameter. There was generally good agreement between the measured S_{11} and the model calculations. The agreement between the measured and calculated S_{12}/S_{11} is less good, however the overall features reproduce qualitatively. The reasons for lack of better agreement are being investigated but probably arise from a combination of wall effects from the tank and uncompensated polarization changes in the biprisms. These measurements are being repeated using horizontal scans in air with several laboratory aerosols to reduce the uncertainties.

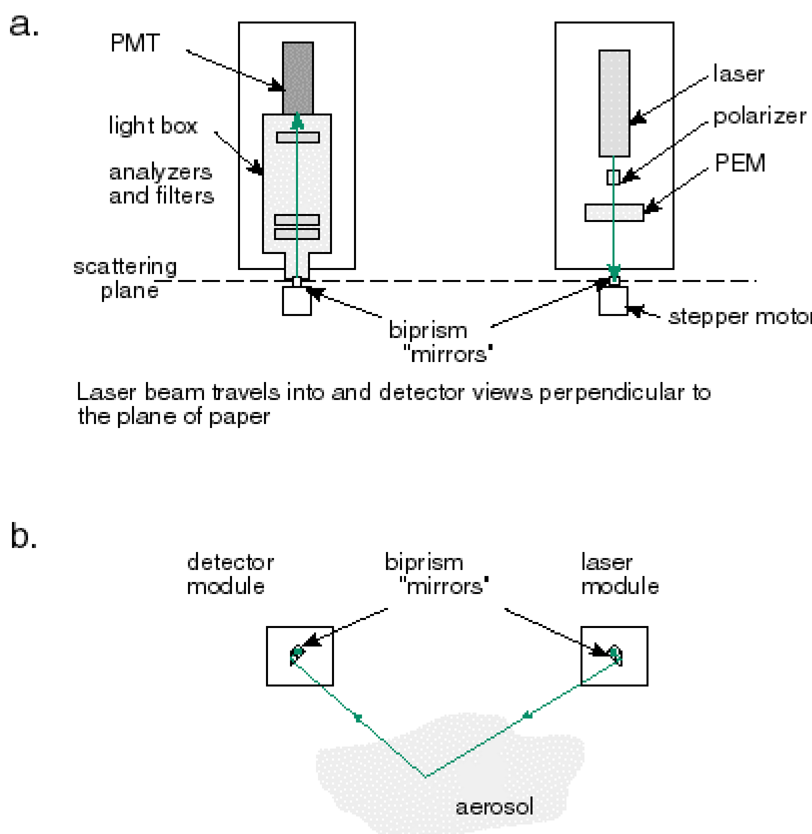

Figure 1. Bistatic Nephelometer as modified, based on original instrument developed by Miller et al. [Refs. 1, 2].

Calculation of Polarized Light Scattering from Soot

To explore the use of the coupled dipole calculation on irregularly shaped particles we examined scattering from soot particles and adapted the computation codes to large scale parallel processing.[4] Soot is an important component of marine aerosols in coastal areas and near shipping lanes with contributions from widespread use of diesel power by shipping, mid-sized pleasure and fishing craft, marine diesel generators, and compressors. This component may have an important effect on the polarized light scattering in the marine atmosphere because the real and imaginary parts of the refractive index of soot are large and variable. This variation is probably due to refractive index determinations based on measurements of soot particles that are not fully dense clusters of primary particles or the presence of homogeneous low- to non-absorptive products in the aerosol. For the purposes of these calculations, several refractive indices were used - real refractive indices were varied from 1.34 to 1.8 and imaginary refractive indices from 0.2 to 0.7.

The CDA is particularly useful for calculating light scattering from soot because the particles can be described as irregular clusters of primary particles.[5] The primary particles are roughly spherical and of the order of 10-20 nm in diameter. For the purposes of these calculations we set the ratio of dipole radius to wavelength to 1:20. If particles this size scatter light of 532 nm, the scattering is close to that described by the Rayleigh equations. Therefore in creating a code to calculate scattering from soot particles, we generated a soot cluster composed of *m* primary particles or dipoles using a random walk routine that we developed.

To explore the effect of this widely-occurring and highly-absorptive component in the marine atmosphere, scattering from a single carbon cluster was calculated using CDA computer codes converted to make use of parallel architecture (Fig. 2 and Ref. 4). The radius of gyration of the soot cluster was roughly 150 nm and the wavelength 532 nm. At this wavelength, based on measurements of graphitic carbon, the refractive index of the primary particles was estimated to be $1.8+0.7i$, therefore initial calculations of scattering used this value. [All the Mueller matrix elements, except S_{11}, have been normalized by S_{11}, the total intensity; the normalized matrix elements, S_{xy}/S_{11} are represented as $\boldsymbol{S}_{xy}$.] Such a calculation shows that scattering from this random cluster clearly could not be described by a Mie computation - this is most obvious from the strong deviation of $\boldsymbol{S}_{22}$ from 1 and $\boldsymbol{S}_{14}$ from 0.

Scattering from a single nonmoving particle would not be observed in the atmosphere. Therefore, to better simulate the random orientation of an ensemble of irregular particles, we first averaged scattering by a single particle which was rotated in space. The particle was rotated at 5^o spacing about the Euler angle ψ [as defined in Ref. 6]. It was then rotated about θ, using $\pi/36$ divisions of θ in cosine space. The results (Fig. 3) show that $\boldsymbol{S}_{12}$ is not equal to -1 at 90^o; $\boldsymbol{S}_{22}$ drops from 1 at angles greater than 90^o; $\boldsymbol{S}_{34}$ is not equal to 0 in the extreme forward direction; and $\boldsymbol{S}_{44}$ approaches but never reaches -1 as the scattering angle approaches 180^o.

The results are exciting for they predict qualitatively several of the observations made on scattering from diesel exhaust where similar particles are observed.[7] Further detailed comparisons of the effects of refractive index, shown in fig. 3, with experimental data reveal interesting features. The backscatter results for $\boldsymbol{S}_{34}$ calculated with a low refractive index are qualitatively more similar to the experimental results than when they are calculated at a high refractive index. Presumably were a greater number of orientations included in the calculations the element would approach 0.

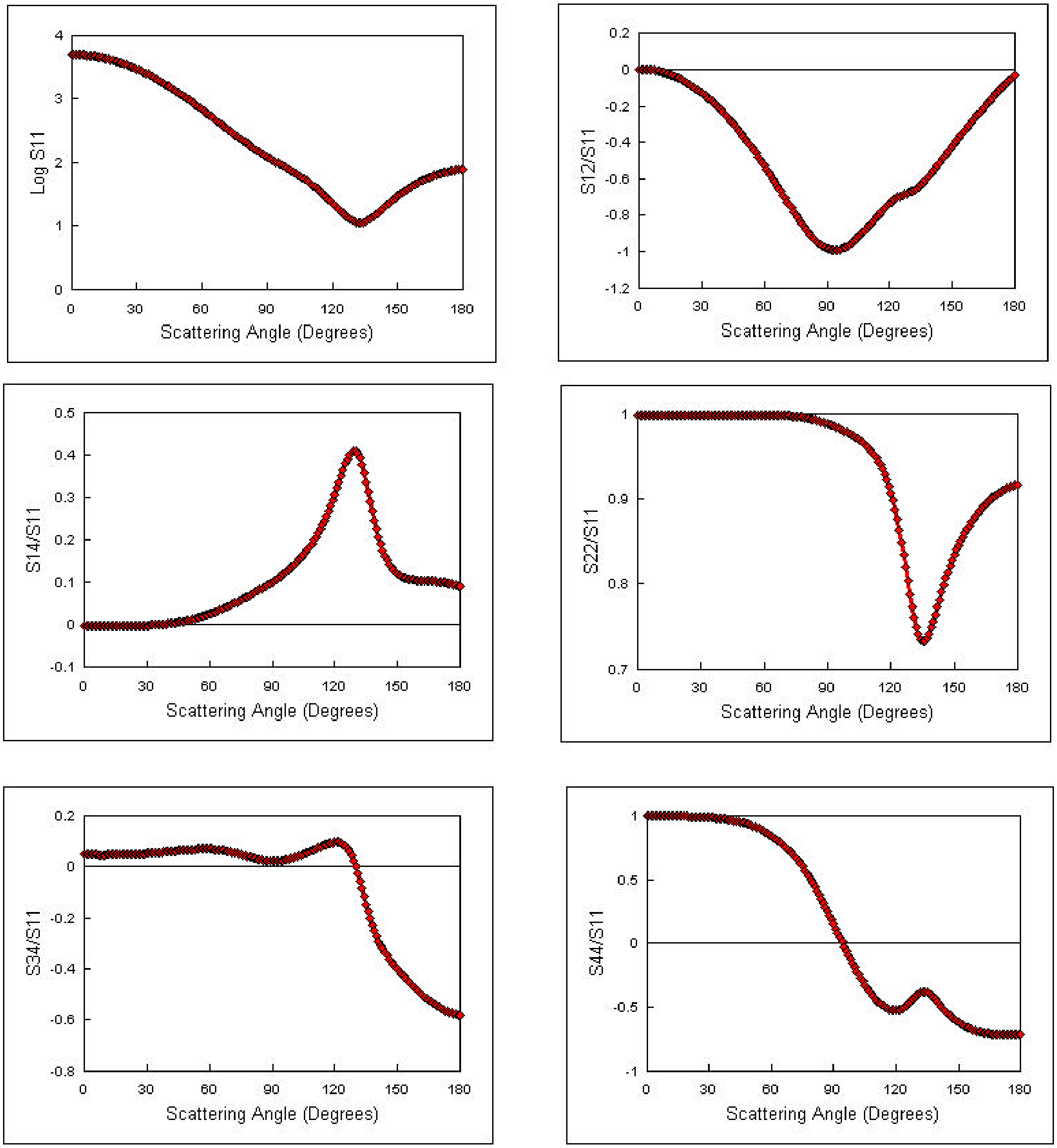

Figure 2. Scattering from a 250-dipole cluster, with radius of gyration ~ 0.1 μm and refractive index for each dipole of 1.8+0.7i.

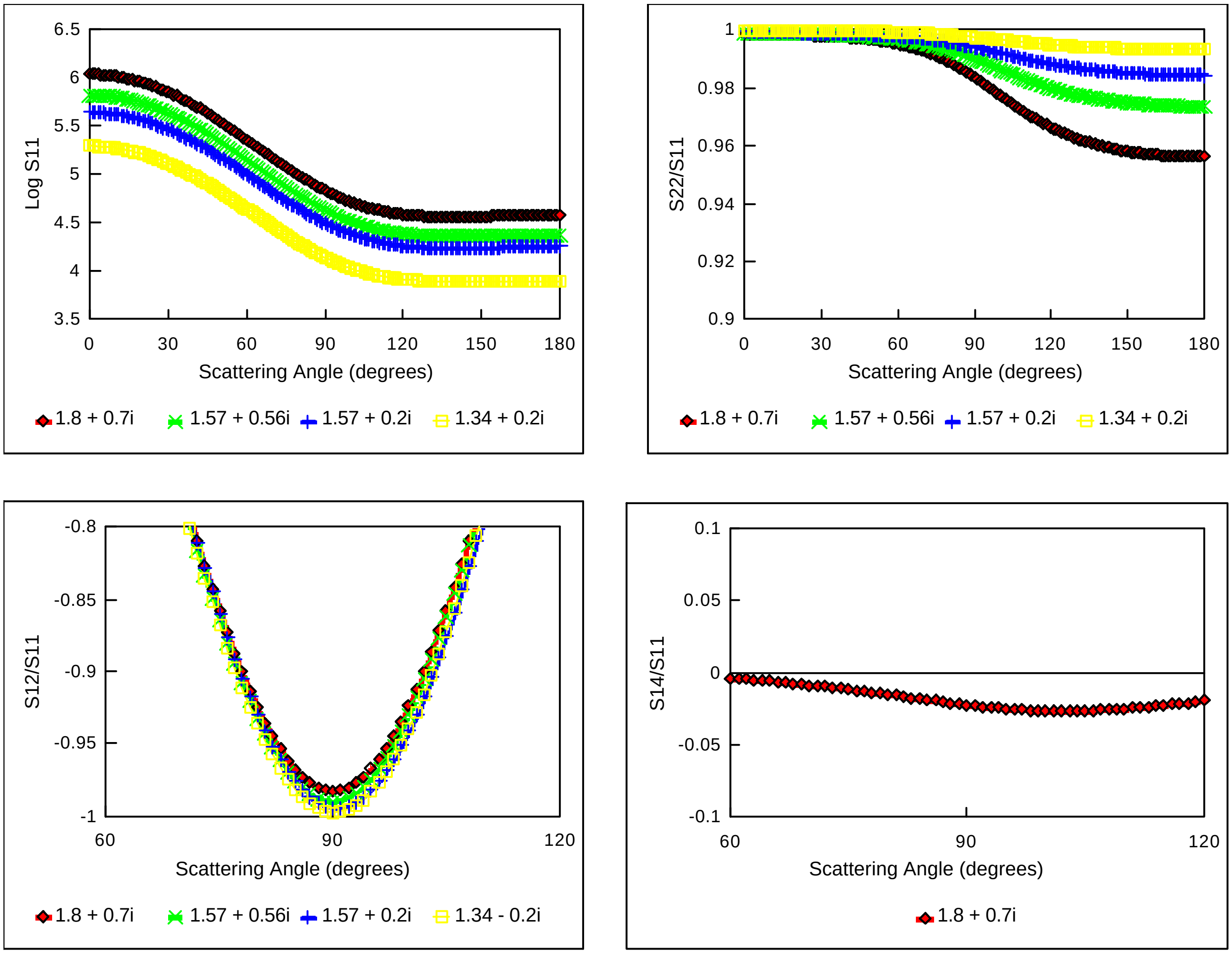

Figure 3. Calculations of scattering from the 250-dipole cluster over all angles using a variety of refractive indices. S_{14} was only calculated at the greatest complex refractive index.

RESULTS

The bistatic polarization nephelometer was modified for the measurement of atmospheric aerosols in the marine atmospheric boundary layer. Polarization modulation and intensity normalization techniques are very well suited for deriving *in situ* scattering properties at a distance. The nephelometer was tested with a water suspension of latex spheres and gave reasonable agreement with theory and further work on verifying its operation with laboratory-generated aerosols are in progress. The instrument should be ready for field measurements in the near future.

Calculations using the CDA adapted to parallel operation show great promise in predicting and modeling the scattering from non-spherical particles in the marine atmosphere. Converting the computer codes to take advantage of parallel architecture vastly enhances the speed of calculation. As the conversion is made more sophisticated, greater enhancement is possible. Calculations of the scattering from a single 250-dipole cluster of soot particles rotated about 2 Euler angles to achieve approximate random orientation reveals all the deviations from the Mie calculations that are actually observed in the scattering from diesel exhaust. As the modeling is refined, greater insights into the nature of the exhausts that are emitted into the marine atmosphere may be derived.

IMPACTS/APPLICATIONS

Non-spherical particles typically have very different polarized scattering properties from that of spherically symmetric particles that require alternative polar enhancement strategies. Thus for an aerosol where non-spherical particles are important, the value of using models based on an assumption of sphericity is questionable and should be carefully evaluated. Deviation from sphericity means that computing the properties of scattered light becomes significantly more complex. Mie scattering calculations can be done using a standard desktop computer. To calculate scattering from all but the simplest non-spherical compact shapes requires a far more powerful computer and extensive time. Adapting CDA codes to parallel structure will greatly enhance the speed of the calculation.

Deviation from sphericity causes changes in the intensities and polarization of scattered light that may be important in improving image contrast. A simple, rapid test which indicates whether such models would be valuable for those using such models to establish the visibility in a particular situation. Such a measurement, based on deviation of $\boldsymbol{S}_{22}$ from 1 is easily made using bistatic nephelometry.

To summarize, because the marine atmospheric boundary layer aerosols, and therefore their polarization properties, are highly variable, it is important to understand that variation in order to optimize polarimetric sensing and imaging systems. By developing a technique to quantify the polarization properties of aerosols in the MABL, it will be possible to make measurements to provide a more quantitative understanding of the effect of polarization on imaging techniques. Measurements will establish the significance of the nature of the aerosols to visibility and image enhancement.

TRANSITIONS

Scanning polarization-modulation nephelometry is presently being developed for use to measure the particulate matter in diesel engines and to determine fiber orientation in the paper industry.

RELATED PROJECTS

We continue with the very fruitful collaboration with Prof. Patricia Hull of Tennessee State University. She spent the summer in Berkeley and was instrumental in developing models of particles with regular shapes and in the initial implementation of the CDA codes on Cray vector processors. We are presently involved in designing and building a scatterometer to measure the exhaust from diesel engines. The scatterometer uses the same polarization modulation and phase sensitive detection used in bistatic nephelometer but is specialized for rapid *in situ* measurements of diesel exhaust stream. In the scatterometer, instead of scanning with time to get the angle dependence, the angle dependence will be measured nearly simultaneously using a multiplicity of detectors. We have also used polarization nephelometry to determine fiber orientation in paper webs for the fiber industry.

REFERENCES

1. Miller, D., Quinby-Hunt, M.S., and Hunt, A.J (1997) Laboratory Studies of the Angle and Polarization Dependent Light Scattering in Sea Ice. *Applied Optics*, **36**, 1278-1288.

2. Miller, D., Quinby-Hunt, M.S., and Hunt, A.J (1996) A Novel Bistatic Polarization Nephelometer for Probing Scattering Through a Planar Interface, *Rev. Sci. Instrum.*, **67**, 2089-2095.

3. Hunt, A.J., Quinby-Hunt, M.S, and Stevens P., (1998) Determination of marine aerosol properties using a bistatic nephelometer, submitted to Ocean Optics XIV, Kona, Hawaii, 10-13 November, 1998.

4 Quinby-Hunt, M.S., Hull, P.G., Shepherd, I.G., and Hunt, A.J.(1998) Calculations of light scattering from non-spherical aerosol particles with parallel processing. Submitted to Ocean Optics XIV, Kona, Hawaii, 10-13 November, 1998.

5. Lahaye, J. and Prado, G. (1981) Morphology and internal structure of soot and carbon blacks. In *Particulate Carbon*, Siegla, D. and Smith, G. eds., Plenum, NY, pp. 33-55.

6. Goldstein, H. (1959) *Classical Mechanics*. Addison-Wesley, Reading, MA. p. 107f

7. Hunt, A.J., Quinby-Hunt, M.S., and Shepherd I.G. (1998) Diesel Exhaust Particles Characterization by Polarized Light Scattering. SAE Paper No. 982629.